

Citation for published version:

Zeidler, A, Salmon, PS, Fischer, HE, Neufeind, JC, Simonson, JM, Lemmel, H, Rauch, H & Markland, TE 2012, 'Reply to Comment on "Oxygen as a Site Specific Probe of the Structure of Water and Oxide Materials"', *Physical Review Letters*, vol. 108, no. 25, 259604. <https://doi.org/10.1103/PhysRevLett.108.259604>

DOI:

[10.1103/PhysRevLett.108.259604](https://doi.org/10.1103/PhysRevLett.108.259604)

Publication date:

2012

Document Version

Publisher's PDF, also known as Version of record

[Link to publication](#)

A. Zeidler, P. S. Salmon, H. E. Fischer, J. C. Neufeind, J. M. Simonson, H. Lemmel, H. Rauch, and T. E. Markland. *Physical Review Letters*, vol 108, article 259604, 2012. Copyright 2012 by the American Physical Society

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Zeidler *et al.* Reply: In our Letter the method of neutron diffraction with oxygen isotope substitution was introduced to measure the structure of light (H_2O) versus heavy (D_2O) water, thus minimizing inelastic scattering effects [1]. Independently, path integral molecular dynamics (PIMD) simulations were made by using various models of water. Best agreement with experiment was found with the simulations made using the flexible anharmonic and polarizable TTM3-F model [2], supporting the hypothesis of competing quantum effects in water where its structural and dynamical properties are governed by an offset between intramolecular and intermolecular quantum contributions [3]. Soper and Benmore [4] object to a remark we make in Ref. [1] on their use of the Empirical Potential Structure Refinement (EPSR) method to model the diffraction data for water shown in Ref. [5] where information is reported on both the intramolecular and intermolecular correlations.

In their Comment, Soper and Benmore state that the results in Ref. [5] do originate directly from measured data sets. This is not, however, the case since in that work only two measured diffraction patterns were used (from neutron and high energy x-ray experiments) and hence the task of determining the partial pair distribution functions $g_{\alpha\beta}(r)$ was under constrained. It was therefore necessary to provide additional information from the reference potential energy model used in the EPSR procedure [5]. In this procedure, the reference model is changed by the introduction of empirical pair potential terms in order to generate revised atomistic configurations for which the calculated structure factors are in better agreement with the measured data sets [6]. Use is made of a feedback (or “confidence”) factor that weights the results either towards those obtained from the reference potential model, such that the extracted pair correlation functions are essentially those of the reference model, or towards the experimental data, such that the extracted pair correlation functions contain more information from the experimental results [6,7]. Hence, while Soper and Benmore’s method based on introducing data from models using EPSR might lead to “quite similar conclusions,” the pair correlations in their work do not come directly from the experimental data but from a procedure which mixes in some modeling data. Indeed this is emphasized by the fact that rather than just using the SPC/E fixed charge model for water, they had to modify it to include an additional exponential term aimed at softening the interactions to improve agreement with experiment [8]. If such tweaks to the input modeling data are needed to improve fits to measured functions then this demonstrates some sensitivity to the model as we refer to in our work.

As shown in Fig. 1 of Ref. [4], the agreement found in reciprocal space between our measured first-difference functions $\Delta F_D(Q)$ and $\Delta F_H(Q)$ and those obtained from EPSR is at first sight satisfactory. Closer inspection shows, however, an underlying dephasing in the high Q range which manifests itself in real space. For example, the intramolecular

peak in the first-difference functions $\Delta G_D(r)$ and $\Delta G_H(r)$ gives O-D and O-H bond distances of $r_{\text{OD}} = 0.985(5)$ Å and $r_{\text{OH}} = 0.990(5)$ Å from our work [1] while “EPSR fits” to the measured time-of-flight neutron diffraction data sets at large Q [5] lead to values of $r_{\text{OD}} \approx 0.971$ Å and $r_{\text{OH}} \approx 1.005$ Å, corresponding to bond length differences of $\approx 0.5\%$ and $\sim 3\%$, respectively. The bond length difference that we find is comparable to the value of $\approx 0.4\%$ found for both ice-Ih [9] and the vapor phase of water [10]. In comparison, the bond length difference from EPSR is much larger than found in any theoretical prediction, unless there are large uncertainties on the EPSR values. For example, different starting points for EPSR referred to in Ref. [4] all involve neutron diffraction data sets for H_2O and D_2O but deliver mean intramolecular bond distances of 0.984 Å [8] and 0.976 Å [11] which are comparable to the O-D distance reported in Ref. [5] but not to the O-H distance.

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Received 31 May 2012; published 22 June 2012

DOI: [10.1103/PhysRevLett.108.259604](https://doi.org/10.1103/PhysRevLett.108.259604)

PACS numbers: 61.05.fm, 61.20.Ja

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